

DOCKET NO: 251072US99RD

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
YUUSUKE SATO, ET AL. : EXAMINER: LEWIS, BEN
SERIAL NO: 10/810,715 :
FILED: MARCH 29, 2004 : GROUP ART UNIT: 1795
FOR: FUEL CELL SYSTEM :

DECLARATION UNDER 37 C.F.R. §1.132

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

I, Yuusuke Sato, state that:

1. I am a graduate of a postgraduate degree of University of Tokyo and received a master's degree in the year 1987.
2. I have been employed by Toshiba, for 20 years as a researcher in the field of chemical engineering such as chemical vapor deposition process and fuel cell system development.
3. I understand the English language or, at least, that the contents of the Declaration were made clear to me prior to executing the same.
4. I am familiar with the above-identified application and know that the current claims, illustrated by Claim 1, define a fuel cell system comprising: a fuel tank storing a fuel comprising dimethyl ether, water, 5-10 wt% of methanol, the mixing ratio of dimethyl ether and water is in a range of 1:3 to 1:4; a vaporizer configured to vaporize the fuel; a reformer configured to reform the vaporized fuel into a hydrogen rich gas; a CO gas removal apparatus

configured to remove CO gas in the hydrogen rich gas; and a fuel cell unit configured to generate electricity by electrochemical reaction of the hydrogen rich gas and oxygen.

5. I understand that the U.S. Patent Office has rejected the claims of this application as being obvious in view of the combination of U.S. 2002/0182460 (Okamoto), U.S. 6,777,116 (Muller) and U.S. 2004/0110046 (Pan).

6. The following experiment was carried out by me or under my direct supervision and control.

7. Presented below are data demonstrating that the concentration of methanol at 5-10 wt% is better than methanol concentrations less than 5wt% and greater than 10% by experiment and calculation.

(1) About methanol concentrations less than 5wt%

(a) In the experiment, DME (46.4mL) and water (15mL) were mixed. Consequently, DME (37.8mL) and mixed solution of H₂O/ DME (20mL) were obtained. In a DME-H₂O system (MeOH-free system), 0.150mol of DME only dissolved in 1mol of water. However, the obtained matter was not suitable for the fuel for the claimed fuel system because the obtained matter immediately separated into two phases (liquid/gas phases).

(b) DME (22mL) and water (22ml) including methanol of 5wt% were mixed. Consequently, H₂O/ DME mixed solution that is in a liquid phase (and homogenous phase) was obtained. In such a case, 0.240 mol of DME dissolved into 1mol of water. The fuel ratio of DME: H₂O was 1: 4.15. Thus, the fuel ratio of DME: H₂O > 1: 4 was achieved. It is also confirmed that all DME (22mL) was stably dissolved in water.

(2) About methanol concentrations greater than 10wt%

As described above, if methanol is added into DME/water solution, the dissolubility to H₂O of DME is improved.

However, if methanol is added greater than 10wt%, the energy density for each unit area of the fuel is decreased.

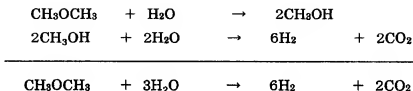
Table 1 shows the mutual solubility of DME and water in a case that DME (22mL) and solution (22ml) of methanol and H₂O were mixed. In Table 1, the concentration of methanol was changed to 5%, 10%, and 20%.

Table 1 DME-H₂O Mutual solubility test

No	MeOH wt%	H ₂ O wt%	DME ml	MeOH水溶液の比重 g/ml	ml-DME/g-H ₂ O	g-DME/g-H ₂ O	mol-DME/mol-H ₂ O	mol-H ₂ O/mol-DME
1	5	95	22	0.98	0.92	0.617	0.242	4.140
2	10	90	22	0.87	1.15	0.766	0.300	3.338
3	20	80	22	0.95	1.32	0.879	0.344	2.906

"MeOH 水溶液の比重" = specific gravity of MeOH solution

In a reform type fuel cell system such as claimed fuel system, in a stoichiometric chemical reaction, 1 mol of DME reacts with 3 mol of water and 6 mol of hydrogen is generated.



According to the Table 1, if 20 wt% of methanol is added, the amount of H₂O/ DME (mol/ mol) is less than 3mol (2.906mol). As it is understood from the equations, the amount of H₂O is stoichiometrically insufficient. 0.1 mol of water may be further added to the fuel tank. However, the volume of the fuel is increased. Accordingly, energy density for each unit area of the fuel is decreased.

Therefore, it is necessary to have the fuel comprising dimethyl ether, water, and 5-10 wt% of methanol, the mixing ratio of dimethyl ether and water is in a range of 1: 3 to 1: 4, to achieve stably generated electricity in a claimed system.

8. I also disagree with the Examiner's conclusion that the claims would have been obvious in view of the combination of Pan, Okamoto and Muller. It should be now

understood from the data above that the concentrations of methanol below 5% yielded a two-phase composition. While I recognize that Pan describes a range of 2-5% touching on the claimed lower value of methanol in the claims, this concentration would while operable in Okamoto's fuel cell system would make Okamoto's system inefficient and impractical.

9. As shown in paragraph [0006] of Pan, Pan considers a DMFC system and describes that 92.5% of water becomes "dead weight" because one water molecule is consumed with each methanol molecule in the electrochemical reaction.

10. When Pan's fuel is supplied to Okamoto's reformer, only 5wt% of methanol would be reformed and hydrogen generated by the methanol (92.5 wt% of water is exhausted). In such a case, the hydrogen generated in Okamoto's reformer would be too small to generate electricity because Pan's fuel only includes 3-5% of methanol. Therefore, it is my opinion that Pan's fuel would not be operable in Okamoto's fuel cell in practice. I have the same view about Muller's applicability in Okamoto's fuel cell as well.

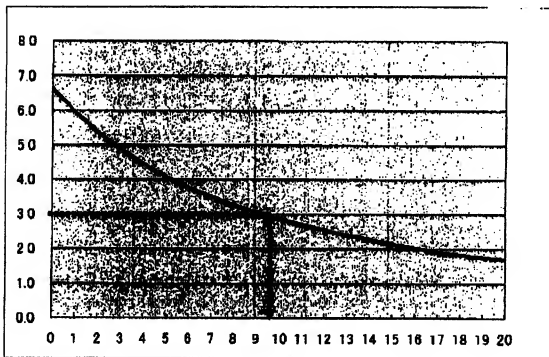
11. In contrast, the claimed fuel which uses DME and methanol for generating hydrogen generates about 67-77 mol% of hydrogen. Thus, the amount of generated fuel is completely different.

12. Therefore, it is my view that the combination of Pan, Muller, and Okamoto do not describe the claimed fuel cell, including fuel nor the effectiveness of what is achieved by that fuel cell and fuel.

13. With respect to the concentrations greater than 10%, while the comparison of methanol for the comparison was two-times the amount of the claimed range and the lower point of comparison in Table 1 of 10%, this comparison is relevant and I am of the opinion that this shows the advantages of the claimed fuel cell system and fuel contained in that system.

14. Further, in a reformed type fuel cell system, it is important that the stoichiometric mixture ratio of DME/water is kept over 3. This is because if the stoichiometric mixture ratio is less than 3, a large amount of carbon monoxide is generated because of the lack of water. As a result, the carbon monoxide poisons the catalyst in the fuel cell thereby decreasing the power generating efficiency in the system.

15. The following graph shows stimulation data demonstrating the relationship between the methanol concentration [wt%] and the mole ratio of DME/water [-].

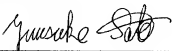


16. If the concentration of the methanol is about 9.6 wt % (about 10wt%), the stoichiometric mixture ratio of DME/water will be over 3. In such a condition, the fuel cell system is stably operated and/or higher power generation efficiency would obtained.

17. On the other hand, when the concentration of the methanol is about 11-12 wt%, the stoichiometric mixture ratio of DME/water is less than 3. This will result in carbon monoxide that will poison the catalyst and/or decrease the power output.

13. As the solubility of DME to water may be changed by the conditions, the fuel having DME, water and 5-10 % methanol with a mixing ratio of DME and water in the range of 1:3 to 1:4 has been determined by me and my colleagues to achieve stably generated electricity. As this is not at all described or suggested by Pan, Okamoto, and Muller, I would not have expected that this particular range of methanol and mixing ratio of DME and water would work as well as it did, which has been shown by the data presented in the application and above in this Declaration.

14. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.



Signature

Oct. 30, 2008

Date